PATENT ABSTRACTS OF JAPAN

(11)Publication number : 10-255823

(43)Date of publication of application : 25.09.1998

(51)Int.Cl. H01M 8/02
H01M 8/10

(21)Application number : 09-053506

(22)Date of filing : 07.03.1997

(71)Applicant : ASAHI GLASS CO LTD

(72)Inventor : YANAGISAWA EIJI
YOSHITAKE MASARU
YOSHIDA NAOKI
ISHIZAKI TOYOAKI

(54) SOLID HIGH POLYMER FUEL CELL

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a fuel cell which is excellent in shape stability and airtightness and can be easily fabricated by constituting a separator of a layer containing a conductive carbon material on a base body surface by using metal containing a specific quantity of aluminium or titanium as a base body.

SOLUTION: Since metal which is elastically deformable and plastically deformable and is excellent in tenacity and contains aluminium or titanium by 80wt.% or more, preferably, 90 to 98wt.% is used as a base body of a separator, the base body can endure mechanical impact since stress is relieved by elastic deformation or plastic deformation even when the stress is applied from the easily collapsing direction. Electric conductivity is imparted by a layer containing a conductive carbon material formed on a base body surface. Therefore, an inexpensive solid high polymer fuel cell having a separator easy to recycle can be obtained.

LEGAL STATUS

[Date of request for examination]

[Date of sending the examiner's decision of rejection]

[Kind of final disposal of application other than the examiner's decision of rejection or application converted registration]

[Date of final disposal for application]

[Patent number]

[Date of registration]

* NOTICES *

1.This document has been translated by computer. So the translation may not reflect the original precisely.
2.**** shows the word which can not be translated.
3.In the drawings, any words are not translated.

CLAIMS

[Claim(s)]
[Claim 1] The polymer electrolyte fuel cell which uses as a base the metal which contains aluminum or titanium 80% of the weight or more, and is characterized by having the separator which comes to form the layer containing a conductive carbon material in this base front face.
[Claim 2] The polymer electrolyte fuel cell according to claim 1 whose layer containing the above-mentioned conductive carbon material is the metallic film which distributed the conductive carbon material.
[Claim 3] The fuel cell according to claim 1 or 2 with which the above-mentioned conductive carbon material has graphite structure.

[Translation done.]

* NOTICES *

1.This document has been translated by computer. So the translation may not reflect the original precisely.
2.**** shows the word which can not be translated.
3.In the drawings, any words are not translated.

DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Field of the Invention] This invention relates to a polymer electrolyte fuel cell.
[0002]
[Description of the Prior Art] Since the polymer electrolyte fuel cell whose operating temperature is about 150 degrees C from a room temperature is excellent in the output characteristics, application in an automobile etc. is expected. Development of the fuel cell cel from which high energy effectiveness and high power density are obtained also for a fuel and the high service condition of an air utilization rate is demanded towards utilization of the above-mentioned fuel cell.
[0003] In a polymer electrolyte fuel cell, a separator is used as a diaphragm between the cells for preventing mixing of the gas of the fuel electrode in the case of carrying out the laminating of two or more cells, and the gas of an air pole. Therefore, it can sometimes machine [excelling in the corrosion resistance and the oxidation resistance in the inside of having conductivity, that gas permeability is small, a lightweight thing, and the high-temperature-steam ambient atmosphere to about 150 degrees C which operates a fuel cell as a property required of the above-mentioned separator,].
[0004] Although carbon system bulk materials, such as an artificial graphite and glassy carbon, are known as a conventional separator ingredient, since a carbon system ingredient is lacking in toughness and weak against it, when it is used as a separator under the conditions in which stress other than a mechanical shock, vibration, and compressive stress exists, the following troubles produce it.
[0005] That is, they are problems, like the energy expenditure required in case the separator which the separator itself is destroyed and cannot maintain a configuration, which a crack arises and cannot maintain airtightness, and which mechanical fabrication is difficult compared with a metallic material, and processing cost is high, and is hard to recycle is manufactured from a raw material is large.
[0006]
[Problem(s) to be Solved by the Invention] This invention maintains a configuration and an airtight, also when stress other than a mechanical shock, vibration, or compressive stress exists, and they tend to carry out fabrication, and the polymer electrolyte fuel cell which has the separator which is industrially practical by low cost is offered.
[0007]
[Means for Solving the Problem] When it is used as a separator for polymer electrolyte fuel cells by the environmental condition in which stress other than a mechanical shock or compressive stress exists, this invention persons can do maintenance of a configuration and a hermetic seal, tended to do fabrication, and in order an assembly is also easy and to obtain a low cost thing, as a result of repeating research variously, they came to make this invention.
[0008] That is, the polymer electrolyte fuel cell which this invention uses as a base the metal which contains aluminum or titanium 80% of the weight or more, and is characterized by having the separator which comes to form the layer containing a conductive carbon material in this base front face is offered.
[0009]
[Embodiment of the Invention] The polymer electrolyte fuel cell of this invention is typically equipped with two or more electrode-film zygotes which consist of an electrode joined by both sides of an electrolyte membrane and this film, and two or more separators. When manufacturing the electrode-film zygote in the fuel cell of this invention, once forming an electrode in the shape of a layer on base materials, such as the approach of forming an electrode directly on the ion exchange membrane which is

a solid-state polyelectrolyte, and carbon paper, various approaches, such as the approach of joining this to an ion exchange membrane or the approach of forming an electrode on [another] monotonous and imprinting this to an ion exchange membrane, are employable.

[0010] As the formation approach of this gas diffusion electrode, although a gas diffusion electrode is used as an electrode in this invention, for example, if the need is accepted further, it mixes with an ostomy agent, a thickener, a diluent solvent, etc., and the catalyst powder which supported the platinum metal to activated carbon etc., ion exchange resin and water repellent, and the existing approach of spraying and applying that mixed liquor to conductive porous bodies, such as an ion exchange membrane or carbon paper, can be applied.

[0011] When an electrode is formed separately from ion exchange membrane, as a conjugation method to the ion exchange membrane of an electrode, hot pressing, the pasting-up method (JP,7-220741,A, JP,7-254420,A), etc. can be applied.

[0012] In the polymer electrolyte fuel cell of this invention, as the quality of the material of the ion exchange resin with which the ion exchange membrane which is a solid-state macromolecule, and the above-mentioned gas diffusion electrode are formed Fluorocarbon sulfonic acid type ion exchange resin is desirable, and they are CF2 =CF2 and CF2 =CF-(OCF2 CFX) m-Op-(CF2) n-SO3 H (among a formula) especially. The perfluorocarbon-sulfonic-acid mold ion exchange resin with which the integer of 0-3 and n consist in m, and 0 or 1, and X consist of a copolymer with F or CF3 in the integer of 1-12 and p is desirable.

[0013] The separator in this invention consists of a base which consists of a metal which contains aluminum or titanium 80% of the weight or more, and a layer containing the conductive carbon material formed in the front face.

[0014] The metal which constitutes a base is aluminum, titanium, an aluminium alloy, or a titanium alloy. Aluminum and titanium can carry out [lightweight]-izing, when the laminating of the separator is carried out, since specific gravity is comparatively small, and since they have the toughness which was excellent compared with the carbon bulk material, they can enlarge reinforcement to mechanical load. Especially aluminum is desirable in respect of low cost in the ease of recycle, and the ease of carrying out of machining. The base is excellent in the pure metal or alloy which uses these metals as a principal component.

[0015] When a base consists of an alloy of aluminum or titanium, the following are specifically illustrated. That is, at least one sort chosen from aluminum, magnesium and manganese, silicon, copper, nickel, a lithium, zinc, lead, a bismuth, titanium, and tin of alloys are used, for example, duralumin, an yttrium alloy, silumin, hydronalium, anti koro DARU, etc. are mentioned. Or an anticorrosion alloy like at least one sort chosen from titanium, aluminum and iron, vanadium, molybdenum, manganese, a zirconium, tin, silicon, palladium, and a tantalum of alloys is mentioned.

[0016] In the above-mentioned alloy, the content of the aluminum which is a principal component, or titanium is 80 % of the weight or more, and is 90 - 98 % of the weight preferably. Since the specific gravity of a base becomes large when the above-mentioned content is less than 80 % of the weight, it is not desirable.

[0017] Moreover, in order to give the corrosion resistance of a base, and oxidation resistance to long-term use in a high-temperature-steam ambient atmosphere, what covered the alloy containing nickel, gold, platinum, or them and the alloy which contains an intermetallic compound further can be used for a base front face. moreover, the purposes, such as raising mechanical strength and making specific gravity small, -- the ceramics, AlPt and nickel3 aluminum, and CuAl2 etc. -- a compound may be distributed in a base. What could use the plate-like thing as a configuration of a base, and established the slot which is the passage of the gas of a fuel or an oxidizer in monotonous one side or monotonous both sides can be used.

[0018] As for especially the thickness of the layer which contains a conductive carbon material in this invention, it is desirable that it is 1 micrometer - 0.5mm 0.1 micrometers - 2mm. Since it will become difficult to form the enveloping layer which followed the covering part made into the purpose if the thickness of the above-mentioned layer is smaller than 0.1 micrometers, it is not desirable. Moreover, since the magnitude of the layered product at the time of it becoming difficult to maintain low resistance and forming a layered product will become large if the above-mentioned thickness is larger than 2mm, it is not desirable.

[0019] The layer containing the conductive carbon material which constitutes a separator consists of a metallic film which distributed conductive carbon material independent a coat or a conductive carbon

material preferably. Each can be used, if it is conductivity as the above-mentioned carbon material, and it will not be limited but an insulator like a diamond will especially be removed. Specifically, an artificial graphite, a natural graphite, carbon black, charcoal, etc. are mentioned. Carbon black begins acetylene black and can also use the carbon black by other processes. A conductive carbon material may be used independently, and two or more sorts may be mixed and used.

[0020] Moreover, especially the thing that has graphite structure also in a conductive carbon material is desirable. The fixed crystal face tends to break and the conductive carbon material which has graphite structure has the description of having lubricity, when deforming in the state of pressurization. A sufficient mechanical strength and sufficient airtightness are maintained to the mechanical stress and the compressive load at the time of this joining the separator and film-electrode zygote which have a conductive carbon material layer to a base front face, and forming a layered product, and the mechanical strength of a film-electrode zygote can also be maintained.

[0021] Moreover, the binder of the silver paste which is the binder of organic systems, such as a conductive graphite paste, or the mixture of an organic system and an inorganic system for the purpose of improving the conductivity of the above-mentioned carbon material, or a platinum paste which maintains the configuration of the layer containing a conductive carbon material, and makes handling easy and which is joined to a base front face may be added in the above-mentioned carbon material. The desirable addition of the above-mentioned binder is 0.5 - 20 % of the weight.

[0022] When using the metallic film which distributed the conductive carbon material as a layer containing a conductive carbon material, as a metal which distributes a conductive carbon material, what is excellent in corrosion resistance is desirable, and one or more sorts of metals specifically chosen from gold, platinum, nickel, Lynn, and a tungsten are illustrated. The content of the conductive carbon material in the above-mentioned metallic film has desirable 1 - 90 capacity %, and it is especially desirable that it is 10 - 60 % of the weight.

[0023] The layer containing a conductive carbon material covers the whole surface or some of base front face, and covers the field which touches an electrode preferably. When covering a part of base front face, an enveloping layer is covered with configurations, such as band-like, a line, island shape, and punctiform, on a base flat surface. The above-mentioned enveloping layer may be arranged regularly and may be arranged irregularly. Moreover, when a base has a rib, the layer containing a conductive carbon material should just cover the field which touches the electrode of a rib at least. Moreover, homogeneity is sufficient as the thickness of an enveloping layer, and an ununiformity is sufficient as it.

[0024] The electric conductivity of a conductive carbon material is 102 although it is so desirable that it is large, since it is important practically to suppress the electric resistance as the whole fuel cell low, and to take out bigger energy efficiently. It is 104-106 especially omega-1 cm to more than [1]. It is desirable that it is omega-1cm-1.

[0025] The formation approach of the layer containing the conductive carbon material on the front face of a base material is produced by the thin film forming methods, such as the thick-film forming methods, such as the mechanical approaches, such as the sticking-by pressure method, print processes, a doctor blade method, and a spray method, a CVD method, PVD, and thermal spraying. Moreover, the metallic film which distributed the conductive carbon material can also be formed by the distributed galvanizing method for galvanizing by using a metal as a binder, using a conductive carbon material as a dispersant. Especially the sticking-by pressure method and the distributed galvanizing method are simple, and desirable from the ability to also form adhesion with a base firmly.

[0026]

[Function] Also when this base requires the stress from the direction which is easy to break by using as the base of a separator the metal which contains aluminum or titanium excellent in the toughness in which elastic deformation and plastic deformation are possible 80% of the weight or more, since stress is eased by elastic deformation or plastic deformation, a mechanical shock can be borne. Moreover, conductivity is also given by the layer containing the conductive carbon material formed in the base front face.

[0027]

[Example] Although an example (Example 1, Example 2, Example 3) and the example of a comparison (Example 4) explain this invention below, this invention is not limited to these.

[0028] aluminium alloy (3.5 mm in 150 mm [150 mm by] x thickness) of the alloy number A5056 by which specification be carried out by JIS-H4000 as a separator which constitute the "Example 1" polymer electrolyte fuel cell to both sides of a base, it pierced so that it might become a configuration as

the width of face which have graphite structure indicate 150 mm and a plate-like conductive carbon material (Union Carbide product name: Grafoil) with a thickness of 2 mm to 150 mm and a height indicate them to be to drawing 1 and it fabricated to them. In addition, in drawing 1 , the width of face 3 of the conductive carbon material 1 is 3mm, and the spacing 2 of conductive carbon material 1 is 2mm. This conductive carbon material is arranged to base both sides, and it is 400kg/cm2. What was pressurized and was pasted up on the base by the pressure was produced.

[0029] Titanium alloy whose presentations are 90 % of the weight of titanium, 6 % of the weight of aluminum, and 3 % of the weight of vanadium as a separator which constitutes the "Example 2" polymer electrolyte fuel cell (1.5mm in 150mm[150mm by] x thickness) Covering with a thickness of 30 micrometers was given using the mask which carried out patterning of the conductive carbon by the spatter on the surface of the base.

[0030] separator (3 mm in 150 mm [150 mm by] x thickness) of the configuration showed in drawing 2 and drawing 3 which be produced by machining by JIS-H4000 using the aluminium alloy of the alloy number A5052 by which specification be carried out as a separator which constitute the "Example 3" polymer electrolyte fuel cell what gave nickel plating covering which distributed carbon black on the surface of the base using the nickel Watts bath which distributed the carbon black particle with a crystal particle diameter of 0.001-0.5 micrometers be used. In addition, the width of face 2 of the slot established in both sides of an aluminium alloy 1 in drawing 2 and drawing 3 is [3mm and the depth of flute 4 of the spacing 3 of 2mm, a slot, and a slot] 0.8mm.

[0031] The separator of the same configuration as the aluminium alloy of Example 3 was produced by machining using the conductive artificial graphite as a separator which constitutes the "Example 4" polymer electrolyte fuel cell.

[0032] What joined the gas diffusion electrode to perfluorocarbon-sulfonic-acid mold cation exchange membrane (Asahi Glass [Co., Ltd.] product name: deflection myon) as a [evaluation result] film-electrode zygote was created. 20 separators produced in Example 1 - Example 4 and the 19 aforementioned film-electrode zygotes were arranged by turns, and the fuel cell was assembled.

[0033] It prepared the ten above-mentioned fuel cells at a time, and the seal test was performed, after repeating the drop test and performing it to a laminating side and a perpendicular direction 10 times on the steel plate with a thickness of 5cm of SS400 from height of 50cm. A result is shown in Table 1.

[0034]

[Table 1]

	気密性良好個数	破損個数
例1	10	0
例2	10	0
例3	10	0
例4	1	2

[0035]

[Effect of the Invention] Also when the laminating of the separator is carried out with a film-electrode zygote under the conditions in which stress other than a mechanical shock and compressive stress exists, it excels in configuration stability and airtightness and fabrication nature can offer the polymer electrolyte fuel cell which has the easy separator which low cost tends to recycle, and which is industrially practical.

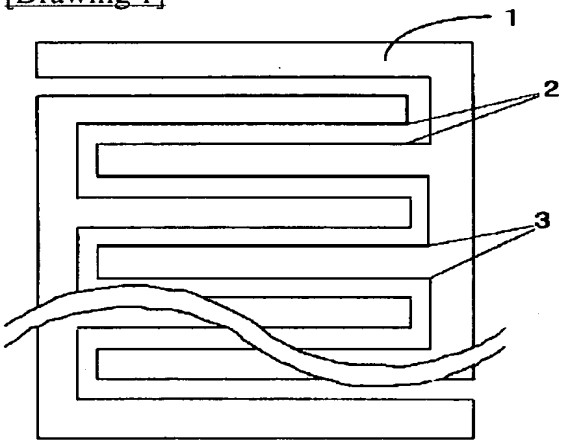
[Translation done.]

* NOTICES *

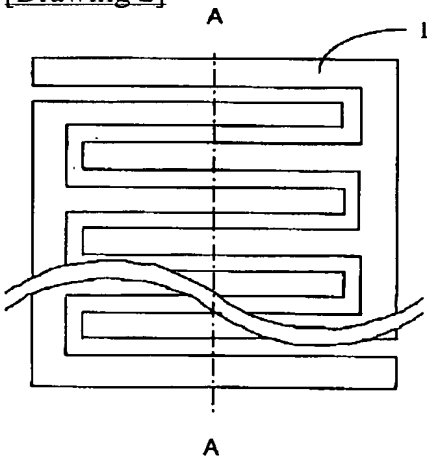
1.This document has been translated by computer. So the translation may not reflect the original precisely.
2.**** shows the word which can not be translated.
3.In the drawings, any words are not translated.

DRAWINGS

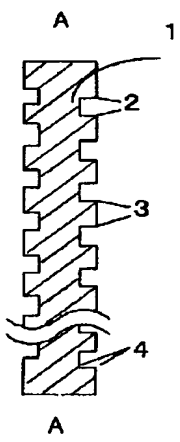
[Drawing 1]



[Drawing 2]



[Drawing 3]



[Translation done.]